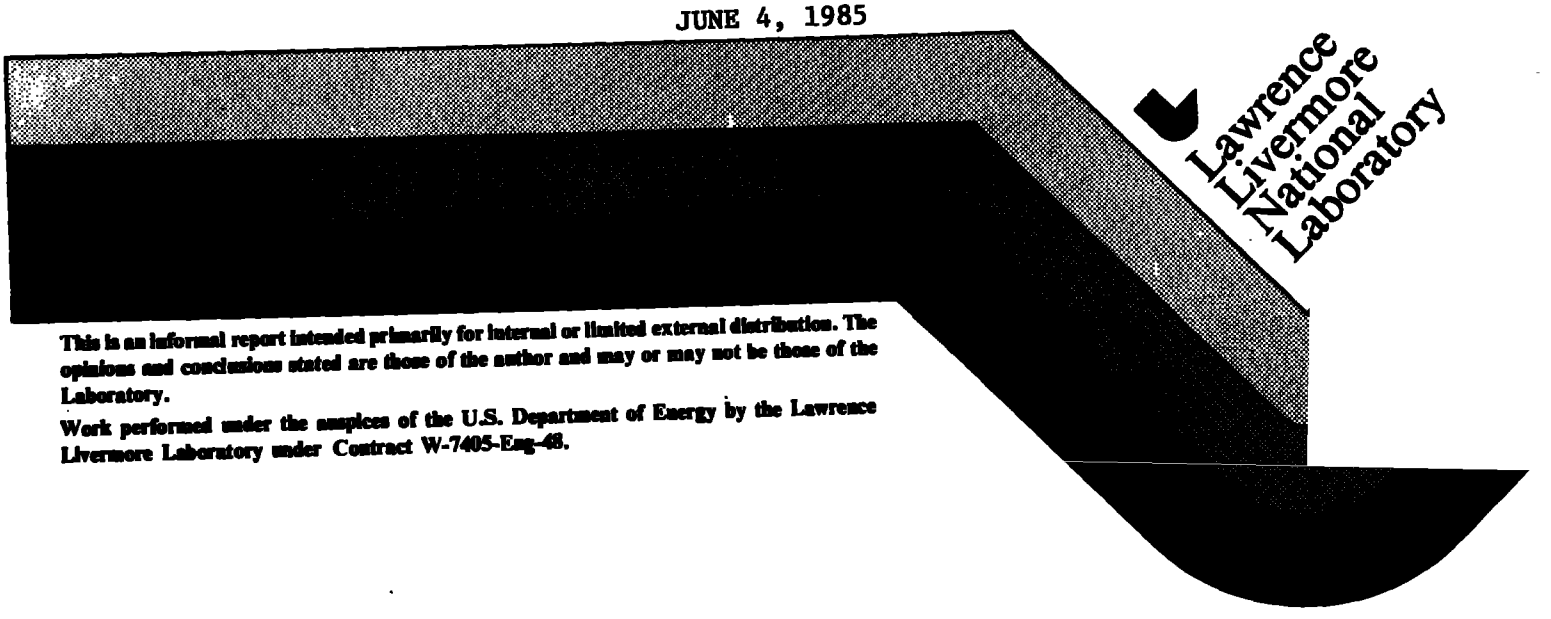
12

UCID- 20381

MARINE SAMPLE EXPLOITATION PROJECT: ESTIMATES REGARDING THE SENSITIVITY AND TECHNOLOGY FOR THE DETECTION OF NUCLEAR EXPLOSION DEBRIS IN THE MARINE ENVIRONMENT

K. V. MARSH
R. W. BUDDEMEIER

JUNE 4, 1985



Work performed under the auspices of the U.S. Department of Energy by the Lawrence Livermore Laboratory under Contract W-7405-Eng-48.

DISCLAIMER

Printed in the United States of America
Available from
National Technical Information Service
U.S. Department of Commerce
5285 Port Royal Road
Springfield, VA 22161
Price: Printed Copy $; Microfiche $4.50

Page Range	Domestic Price	Page Range	Domestic Price
001-025	$ 7.00	326-350	$ 26.50
026-050	8.50	351-375	28.00
051-075	10.00	376-400	29.50
076-100	11.50	401-426	31.00
101-125	13.00	427-450	32.50
126-150	14.50	451-475	34.00
151-175	16.00	476-500	35.50
176-200	17.50	501-525	37.00
201-225	19.00	526-550	38.50
226-250	20.50	551-575	40.00
251-275	22.00	576-600	41.50
276-300	23.50	601-up[1]	
301-325	25.00		

[1]Add 1.50 for each additional 25 page increment, or portion thereof from 601 pages up.

Rationale

We initiated the LLNL Marine Sample Exploitation Project in response to interest in developing a capability to monitor and evaluate clandestine nuclear testing activities in the oceanic regions of the southern hemisphere. The primary focus is on evaluation of compliance with the Limited Test Ban Treaty (LTBT) and the Non-Proliferation Treaty (NPT).

Sampling of marine plankton can be an effective method for detecting and evaluating radionuclides to provide corroborative evidence when a nuclear test has been conducted. Two advantages of such sampling are that marine plankton accumulate many isotopes with concentration factors (relative to water) much greater than 1000, and they can act as a repository of evidence. Hence, collections are not time urgent (in contrast to air sampling), and sampling is simple and can be done in international waters without dependence on intercountry agreements. Once collected, plankton samples can be analyzed to identify the presence of debris unique to nuclear explosions. Plankton sampling has several advantages over water sampling: plankton can be collected very quickly without the need for complicated or expensive apparatus; a large number of radionuclides can be determined immediately by direct counting without further processing; and the samples are easily stored, transported, and conveniently analyzed by wet radiochemical methods if necessary. To a certain extent, other marine organisms (such as algae and grasses) also accumulate isotopes and can be used as data sources. Not much is known about the utility of marine biota for these purposes, and there is a need to demonstrate a capability for marine sampling and analysis as a means of identifying clandestine nuclear events in the ocean.

Purpose

The purpose of this report is to supply some facts, figures, assumptions, and reasonable guesses about the prospect of detecting a small nuclear explosion in which most of the fission products are released to the ocean. While at first glance ocean dispersal might seem to present a formidable dilution problem, the use of marine plankton as a bioaccumulator reduces this problem considerably. Marine plankton have high concentration factors for most of the radionuclides associated with nuclear explosions (Marsh and Buddemeier, 1984). The real questions are, how high is the sensitivity and how difficult is it to locate the contaminated area? This report is an attempt to answer both of these questions.

Sensitivity

The sensitivity question can be addressed by estimating the area over which a given amount of radioactivity can be dispersed and still be detectable. Could we, for example, expect to detect the fission products from a 1-kt equivalent of fission dispersed over tens of thousands of square kilometers? Such an area could be either the result of debris originally distributed over a large area, or an initially small zone of contamination that had undergone radioactive decay and mixing in the ocean over a period of many weeks (or even months). The direct external radiation field in such an area would probably be indistinguishable from background even with sensitive instruments, but concentration by plankton can help considerably.

About 1.45×10^{23} fissions are required to produce a 1-kt explosion; with information from Bolles and Ballou (1959), we can calculate that the residual radioactivity after 7 d is about 3×10^{18} dpm. Several isotopes of the rare earth elements each contribute about 10% of the total radioactivity or

3×10^{17} dpm. Our detection limits for these nuclides are all about 0.05 dpm/g of plankton based on an average 200-g sample (Marsh and Buddemeier, 1984). If we take 10^3 as a conservative average of the concentration factor for rare earth nuclides, a gram of plankton becomes equivalent to a liter of seawater. Thus, we can detect 1-kt of fission products diluted by about 6×10^{18} L or 6×10^{15} m^3. This estimate is based on bulk sample measurements by direct Ge-Li γ-spectroscopy and sensitivity could easily be increased a factor of 10^2 to 10^3 by dissolving the sample and using wet radiochemical analysis techniques. In order to see what 6×10^{15} m^3 implies in terms of ocean surface area, we start by assuming some depth of mixing. Previous experience (Hines, 1962) suggests that the radioactivity will not be uniformly mixed, but will be stratified by the plankton at several depths throughout the mixed layer (~100 m). However, because most of the radioactivity will still be in the water (plankton is only about 0.2 ppm in most ocean water), let us assume that it is uniformly mixed down to 100 m and that our 6×10^{15} m^3 is represented by an area of 6×10^{13} m^2. This is about 1/10 the area of the globe or 1/8 the surface of all the world's oceans combined. Let us take a more realistic situation as a practical example. Say a 1-kt equivalent of fission products is dispersed over 10^5 km^2 (300 x 300 km) and mixed to our previous depth of 100 m for a dilution volume of 10^{13} m^3 or 10^{16} L. The resulting concentration is 30 dpm/L, 600 times our detection limit.

Other estimates of this type can be made assuming values for the parameters of area, nuclear yield, depth of mixing, and fission product age. In the case of a very small area, water samples (rather than plankton samples) might be sufficient to determine that a test had occurred. However, some of the more diagnostically valuable radionuclides would still be in low concentration, and plankton sampling would permit better measurements of

them. The other parameters are subject to various practical constraints. Nuclear yields of much less than 1-kt equivalent are unlikely, but could occur, for example, in the case where most of the fallout is on land with only a small fraction reaching the ocean. A test much larger than about 10 kt would probably be detectable by other means unless it were fired deep underwater. A 100-m depth of mixing is probably about a maximum for all but deep underwater explosions, as it is the average depth of the mixed layer in the ocean. Finally, with regard to an assumed age for the fission products, it would require at least several days to arrive at the site, while after a few months the debris, although probably still detectable, would be more difficult to associate with a recent nuclear explosion because of decay of unique isotopes. Whatever set of parameters is chosen, it is still clear that detection at high dilution is not our principal problem.

Location of the Contaminated Area

Whether the contaminated area is large or small, finding it will be our most difficult problem. If the area is very large and diffuse as discussed above, plankton samples will have to be collected and analyzed on station to provide adequate sensitivity. If the contaminated area is very small and sharply delimited, finding it could be difficult. In either case, we will assume that we know approximately where to look and discuss some techniques that would be applicable.

Let us take the case of a very small spot, say 10^3 km^2. This is a circle roughly 40 km in diameter, one that a ship can circumnavigate in 8 h or so, as was done after the Wahoo underwater test at Enewetak in May 1958 (Hines, 1962). While, for the sake of argument, we will assume a circular spot, it should be noted that this spot will most likely not be even roughly

circular. Observation of dye patches and oil slicks suggests that under the influence of wind and current, the spot usually becomes a greatly elongated ellipse or even a sickle shape. Let us assume that our hot spot of 10^3 km^2 is known to be located somewhere within an area of 10^6 km^2. This is a pessimistically large area to search; knowledge about the origin of the debris, winds, and ocean currents would reduce it by at least an order of magnitude. One technique for finding the spot might be to deploy multiple plankton collectors that would cover the search area in a short time and be recovered by ship or aircraft, perhaps after signalling whether or not a radioactive sample had been collected. If a self-powered (or wind-powered) plankton collector could be developed, a ship could deploy a series of them along a boundary line of the area to be searched. If the 10^6 km^2 area were a square 10^3 km on a side, 25 collectors released 40 km apart would have a high likelihood of intercepting a spot of about 40-km diam within their range. If the range were 100 km, 10 such releases would be required for a maximum of 250 collectors, assuming that none was recovered and used again. Ideally, the collectors would each be equipped with a radiation detector and locator beacon and would transmit information about which of them had radioactive samples and what their locations were. If the speed of the collectors was about 5 km/h (2.7 knots), they could cover the square in about 200 h not including deployment time. By contrast, a single ship would have to travel about 25,000 km to search on a series of 40-km parallels (a little less if an optimum search strategy is used), towing for plankton most of the time. Even a speed of 10 km/h would require 2500 h or 100 d of ship time. At a conservative price of $4,000/d, this is a $400,000 expense, and 100 d allows too much time for further dispersion and decay. If each self-powered collector cost as much as $1,000 and was used only once, the method would be

cost and time effective, as ship time would be reduced to about 25 d and the spot would not disperse very much during the search period. Aircraft deployment and recovery or some combination, such as aircraft deployment and ship recovery, would reduce the time and probably expenses even further, as would collectors capable of a several-hundred- to a thousand-km range. These collectors would also be very useful when searching for a large diffuse zone of contamination.

The spot might also be located by an aerial radiation survey similar to the one flown by EG&G in 1972 as part of the Enewetak radiological survey. By using a large array of sodium iodide detectors mounted in a helicopter flying at 100 ft. above ground level, EG&G was able to measure levels at 1 m above the ground of 1 μR/h above background (Stuart and Meibaum, 1973). Background on the surface of the open ocean is about 3 μR/h, so let us estimate what the level might be in our theoretical 10^3 km^2 hot spot.

From Fleming (1967), the radiation field on land 3 ft above the surface from 1-wk-old fission products is calculated to be 5 R mi^2/h kt, or 13 R km^2/h kt. To convert this to an ocean estimate, we make two assumptions: (1) the average photon energy is 1 MeV, and (2) the radionuclides are uniformly mixed down to 100 m. From Evans (1955), the half-thickness for absorption of 1-MeV photons in water is about 25 cm, so let us say that there is no contribution to the radiation field from nuclides below a depth of 0.25 m and no absorption of photons from 0.25 m to the surface. Multiplying the 13 R km^2/h kt by 0.25/100, we get 0.032 R km^2/h kt, or 0.032 mR/h kt dispersed over 10^3 km^2, well above the 1 μR/h sensitivity of EG&G. While this estimate combines calculations and simplifications, other data from two underwater tests lend some credence to it.

Hines (1962) describes the experience of the ship Rehoboth in its mission

to survey the radioactive area produced by the Wahoo test, which was fired south of Enewetak Atoll in May 1958. At H + 1 h, the ship had no trouble finding a field increasing rapidly to 500 mR/h, and in fact was forced to retreat. If we assume (because this was an underwater explosion) that the mixing was complete after 1 h, and use the well-known $T^{-1.2}$ relationship for the decay of mixed fission products, we can calculate that the field at 1 wk would have been at least 1.1 mR/h, neglecting any further dilution or dispersion. When compared with our lower calculated value of 0.032 mR/h, the agreement is not too bad considering: (1) the calculated value was intentionally very conservative, (2) we neglected dilution and dispersion from the Wahoo test, (3) we do not know the precise area or maximum intensity of the Wahoo spot, and (4) the 0.032 mR/h is for 1-kt yield; while the Wahoo yield is still classified, it was probably somewhat larger than that.

Results of another underwater test, Wigwam, are somewhat more detailed (Joseph et al., 1971). Wigwam was a 30-kt device detonated at a depth of 610 m off the southwest coast of California in 1955. After 6 d, the surface pattern of distribution of radioactivity as determined from an aircraft at 150-m altitude was roughly an ellipse 36 km x 4.5 km ($A \sim 133$ km^2) with radiation contour levels measured from 0.01 to 0.25 mR/h. While these measurements seem to bracket our calculated value of 0.032 mR/h, it must be pointed out that the Wigwam yield was 30 times larger and the area of the spot eight times smaller than we assumed in the calculation. If we reduce the 0.25 mR/h in proportion to 1 kt/30 kt and 133 km^2/1000 km^2 to get a number comparable to our calculated value, we get a maximum of only about 1 μR/h, considerably below our estimate and barely detectable. Furthermore, a spot of this size and shape might very easily escape detection altogether unless a grid pattern about 30 km on a side were used or fairly precise information

were available regarding its location.

It might be argued that the only two experimental results were injections underwater and that a significant fraction (maybe most) of the radioactivity remained below the γ-radiation total absorption depth, resulting in a residual radiation field much lower than would be produced by an equivalent amount of radioactivity deposited from fallout on the surface. If so, then our calculation is more valid than the experimental results, and the hot spot produced by fallout of a few kilotons equivalent of fission products would probably be detectable by sensitive radiation monitoring instruments. Also, regardless of speculation, the fact remains that a 30-kt underwater shot was easily detectable six days later for those who knew where to look. Simple extrapolation suggests that even a 3-kt shot would have been detectable.

If the spot could be located by an aerial survey, rather than a ship survey, certainly time and possibly expense would be reduced. An aircraft flying about 100 mi/h (160 km/h) would require about 160 flying hours to complete the previously discussed 25,000 km-survey track. With sufficient aircraft, the whole survey could be flown in a day or two. If we assume a cost of about $300/flying hour, this comes to around $50,000 for the survey, to which must be added the cost of sampling for probably about 10 d or another $40,000. One can envision a combination of techniques involving an aerial survey and deployment of the samplers followed by ship recovery and further sampling, which could provide information a few days after arrival in the vicinity.

Sensitive equipment such as the EG&G array is delicate and expensive. However, if a very cheap, even "throwaway," floating detector could be built that was capable of responding to fields of a few μR to a few mR and transmitting radiation level and location information, then thousands of these

could be dispersed over the search zone by a land-based fast airplane flying at any altitude up to a few thousand feet. For example, an aircraft flying at 400 knots (740 km/h) and dropping a detector every 10 km (1 detector per 100 km^2) would have to fly 100 times across the 10^3 x 10^3-km search zone for a total distance of 100,000 km or a flight time of 135 h, deploying a total of 10^4 detectors. Even at $5.00 each for the detectors, costs would be $50,000 for detectors and probably at least that much more for flight time. Total time would be only marginally reduced; however, the detection grid would be much finer (10 detectors in our theoretical 10^3 km^2 hot spot), resulting in a very high probability of finding the spot. If we are willing to settle for fewer detectors, or can in some way reduce the size of the search area, then all costs come down proportionately. Besides being simpler than the sodium iodide array, the floating detectors would remain in place and drift with the hot spot as continuous monitors. These detectors could also be used on land as well as sea and might find other applications for radiation field detection and monitoring.

Conclusion

It seems very likely that whether a fission product release results in a very diffuse low-concentration contaminated area, a small concentrated area, or something in between, techniques are available that would ensure a high probability of detection and provide samples for corroborating evidence. The more a search zone can be reduced by use of intelligence and meteorological information, the higher the detection probability will be and the lower the cost.

References

Bolles, R. C., and N. E. Ballou, "Calculated Activities and Abundances of the U^{235} Fission Products," Nuclear Science and Engineering 5 (3), 156 (1959).

Evans, R. D., The Atomic Nucleus (McGraw-Hill Book Company, Inc., 1955), 714.

Fleming, E. H. Jr., The Fission Product Decay Chains (Pu-239 with Fission Spectrum Neutrons), Lawrence Livermore National Laboratory Report UCRL-50243, Vol. I, 6 (1967).

Hines, N. O., Proving Ground: An Account of the Radiobiological Studies in the Pacific, 1946-1961 (University of Washington Press, Seattle), 278 ff (1962).

Joseph, A. B., P. F. Gustafson, I. R. Russell, E. A. Schuert, H. L. Volchok, and A. Tamplin, "Sources of Radioactivity and Their Characteristics," Radioactivity in the Marine Environment (National Academy of Sciences, Washington, D.C., 1971).

Marsh, K. V., and R. W. Buddemeier, Marine Plankton as an Indicator of Low Level Radionuclide Contamination in the Southern Ocean, Lawrence Livermore National Laboratory Report UCRL-53549 (1984).

Stuart, T. P., and R. Meibaum, "Aerial Radiological and Photographic Survey," Enewetak Radiological Survey, W. E. Nervik and R. Ray, Eds., USAEC Nevada Operations Office, Las Vegas, NV, NVO-140, Vol. I (1973), pp. 71-81.